THERMAL WAVE RESONATOR : IN SITU INVESTIGATION BY PHOTOTHERMAL DEFLECTION TECHNIQUE

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ABSTRACT

The photothermal deflection technique applied to a gas thermal wave resonator seems to be one of the most powerful techniques to investigate in situ the thermal diffusivity of the gas. After a brief description of what a thermal wave resonator really is, a discussion is presented of the advantages and disadvantages of its use for measuring the gas thermal diffusivity.

KEY WORDS: air; interference; photothermal deflection technique; thermal diffusivity; thermal wave.

1. INTRODUCTION

Although the thermal wave interferometry [1] has been applied for long time in order to measure the thickness or the thermal diffusivity of thin solids, the name thermal-wave resonator [2] has been introduced only two years ago. Basically the physical process which takes place in a thin film periodically heated at one side is absolutely the same of that one which occurs in a thermal wave resonant cavity: the behaviour may be described using the interference between thermal waves propagating in opposite directions.

A plane thermal wave resonator is an open cavity between two solids samples with plane facets which behave as mirrors. From a thermal point of view this condition is practically always verified. In fact due to the great differences between the thermal effusivities of solids and gases, when a thermal wave propagating in the gas approaches the solid, the temperature rise on the solid is kept to zero because the solid has a large *thermal inertia*, but the thermal gradient reaches its maximum value. So if one measures close to the mirror not the temperature rise, but a quantity proportional to the thermal gradient (*heat flux, photothermal deflection angle*), one finds a double value that means an amplification of the signal. So a first use of a resonant cavity (see Fig.1) is for amplifing a signal related with a thermal flux, as it happens for example in the photothermal deflection technique for a direct measurement of the gas thermal diffusivity.

2. MEASUREMENT OF GAS THERMAL DIFFUSIVITY

Generally in order to measure the thermal diffusivity of non-absorbing gases through the photothermal technique one has to heat a reference solid sample, in contact with the gas, and look at the heat diffusion induced in the gas itself. One of the simplest methods consists of illuminating periodically the sample by the means of a wide spot laser beam (*pump*) so to generate a plane thermal wave in the gas. The oscillating gas temperature depends on the distance from the sample surface z as follows

$$T(z,t) \approx \frac{I}{e_s\sqrt{2\pi f}} \exp\left(-z/l_{gas}\right)\cos\left(2\pi ft - z/l_{gas}\right) \ , \qquad (1)$$

where I is the laser intensity, f is the modulation frequency, $l_{gas} = \sqrt{D_{gas}/\pi f}$ is the gas thermal diffusion length and D_{gas} is the gas thermal diffusivity, while e_S is the sample thermal effusivity. To detect it a second laser beam (*probe*) can be used travelling in the gas at some distances z from the solid. Its deflection induced by the thermal gradient in z (*mirage effect*) is given by

$$\Phi = \frac{l_{path}}{n_{gas}}\left(\frac{dn_{gas}}{dT}\right)\frac{dT(z)}{dz} \qquad (2)$$

where n_{gas} and dn_{gas}/dT are the refractive index and the optothermal coefficient of the gas respectively and l_{path} is the effective length in which the probe beam deflection occurs. By combining Eq.(2) with Eq.(1) and looking at Φ in terms of amplitude and phase one can write

$$\begin{aligned} \ln\left(|\Phi|\right) &= -z/l_{gas} + \ln\left(\frac{l_{path}}{n_{gas}}\frac{dn_{gas}}{dT}\frac{I}{k_s}\sqrt{\frac{D_s}{D_{gas}}}\right) \\ \arg\left(\Phi\right) &= -z/l_{gas} + \theta_0 \end{aligned} \qquad (3)$$

where k_S and D_S are the sample thermal conductivity and diffusivity while θ_O is a constant quantity. Eqs.(3) show that both logarithm of amplitude and phase have the same linear behaviour as a function of z/l_{gas} and suggest two methods to calculate the gas thermal diffusivity:

1) Frequency scan method.

In this method the probe beam travels at a fixed height z, while the frequency of the periocal heating is changed. By plotting the phase and the logarithm of amplitude of the photothermal deflection signal as a function of $\sqrt{f}$ the same slope is obtained from which the diffusivity can be worked out by one of the two relationships

$$D_{gas} = \pi z^2 \left(N_f \Big/ \frac{\Delta \ln(|\Phi|)}{\Delta \sqrt{f}} \right)^2 \qquad (4a)$$

$$D_{gas} = \pi z^2 \left(N_f \Big/ \frac{\Delta \arg(\Phi)}{\Delta \sqrt{f}} \right)^2 \qquad (4b)$$

Note that a corrective factor N_f has been introduced to take into account the effect of the finite spot size *a* of the Gaussian pump beam on the sample surface. The main consequence of a finite spot size is the bending of the plane thermal wave which, leaving the sample surface, tends gradually to a spherical wave. In figure 2 the result of a numerical study on the factor N_f is shown. The N_f values for both amplitude (see Eq.4a) and phase (see Eq.4b) are plotted as a function of the parameter *a/z* for different diffusivity ratios D_{gas}/D_s (0.2 - 0.5 - 5). For large values of *a/z* all curves tend to the value 1, which represents the value obtained in the case of a plane thermal wave. Note also that N_f for the phase stays around 1 for any value of *a/z* while N_f for the amplitude may deviate more from 1. In practice, by choosing a ratio *a/z* around 10 and looking at the expression obtained considering the phase, one does not commit a serious error by putting N_f =1 into Eq.(4b). Indeed the main sources of error for D_{gas} in this method are due to the inaccuracy on *z* and on the calculated slope $\frac{\Delta \arg(\Phi)}{\Delta \sqrt{f}}$. Particular care in fact should be given to align the probe beam (it means to keep *z* constant along the path) and to reduce its spot (that means Δz as low as possible).

2) Height scan method.

In this case the frequency (that is related to the *thermal wavelength*) is fixed, while the probe beam skims the sample surface at different heights *z*. By plotting the data of phase and logarithm of amplitude as a function of *z* , also in this case, the same slopes are obtained, from which the diffusivity can be worked out by one of the two relationships

$$D_{gas} = \pi f \left(N_z \Big/ \frac{\Delta \ln(|\Phi|)}{\Delta z} \right)^2 \qquad (5a)$$

$$D_{gas} = \pi f \left(N_z \Big/ \frac{\Delta \arg(\Phi)}{\Delta z} \right)^2 \qquad (5b)$$

In this case another corrective factor N_Z has been introduced to take into account the effect of *a*. In figure 3 the results of a numerical study on the factor N_Z is shown. The N_Z values for both amplitude (see Eq.5a) and phase (see Eq.5b) are plotted for different diffusivity ratios D_{gas}/D_S (0.2 - 0.5 - 5) as a function of the parameter a/l_{gas} which remains constant during the whole measurement. Also in this case it comes out that the larger is a/l_{gas} the more all curves tend to the value 1 and that only for the phase N_Z is kept around 1 for any value of a/l_{gas} and diffusivity. The suggestion is therefore to apply Eq.(5b) without considering the influence of N_Z (that means to fix N_Z=1). Note that in this case the absolute value of *z* is not requested so that the accuracy of Eq.(5b) is better than the one of Eq.(4b).

3. THERMAL DIFFUSIVITY MEASUREMENT IN A THERMAL CAVITY

Another approach for measuring the gas thermal diffusivity refers to the study of the deflection angle inside a gas thermal wave resonator. In this case the simple thermal wave resonator shown in fig.1 is used. It is made of two plane thermal mirrors and is filled with the gas to be measured. The first plane mirror is a thick glass layer coated by a thin pump-absorbing film (1μm Silicon). The use of the glass on one hand allows to reflect the thermal waves inside the cavity, and on the other hand let the pump beam, coming from outside the cavity, to illuminate directly the thin film. The second plane mirror is a thin aluminium foil (20μm). It is worth to note that this thickness is larger than that needed to behave as a thermal mirror [3].

The probe beam is placed inside the cavity at a distance *z* from the first mirror, which is also the heat source. The theory of thermal wave interferometry applied to this simple system allows one to write for the deflection angle

$$\Phi = \left(\frac{l_{path}}{n_{gas}} \frac{dn_{gas}}{dT} \frac{I}{k_s} \sqrt{\frac{D_s}{D_{gas}}} \right) \frac{\exp\left[-(1+j)z/l_{gas}\right] + R_2 \exp\left[-(1+j)(2L-z)/l_{gas}\right]}{1 - R_1 R_2 \exp\left[-2(1+j)L/l_{gas}\right]} \quad (6)$$

where L is the cavity length, R_1 and R_2 are the thermal wave reflectivity of the two mirrors, usually ranging between *0.99* and *1*. By comparing the deflection in the cavity (Eq.6) with the deflection without cavity (Eq.3 or more simply Eq.6 with the second mirror removed $R_2=0$), one immediately realises that the cavity behaves as a deflection amplifier, which, theoretically, in the case of a thin cavity ($z,L<\lambda_{gas}$) has a gain $(1+R_2)/(1-R_1R_2)$ that is a large quantity considering that $R_1,R_2 \cong 1$. In practice the gain decreases when one considers the effect of the finite pump spot size a. In fact the heating, in this case, produces plane thermal waves propagating not only along the cavity but also in undesired directions, determining in such way a loss of amplification.

Although Eq.(6) guarantees a stronger signal, the amplitude and phase signal have a complex behaviour so that no easy way seems to exist, except a nonlinear fit, to calculate the gas thermal diffusivity.

A different method is here considered, obtained by a different way to look at Eq.(6). The deflection, in fact, can be seen as the sum of two terms which refer to the forward and backward thermal waves. It is well know that the ratio between the backward and the forward wave, that is the *reflection coefficient*, has an easy single-exponential expression useful for a linear fit. One may therefore ask if it is possible, starting from the sum of the two quantities mixed together in Eq.(6) to calculate their ratio. The answer is positive, if an additional information is provided, as for example the value of the forward wave when the backward wave is absent. In other words to calculate the reflection coefficient Γ, the deflection Φ has to be compared with its value obtained without cavity or when the cavity length L is made to tend to infinity. The coefficient Γ is then given by [4]

$$\Gamma(L) = \frac{\Phi(L) - \Phi(\infty)}{\Phi(L) + \Phi(\infty)} = \frac{R_2(1+R_1)}{2} \exp\left[-2(1+j)(L-z)/l_{gas}\right]$$

So that

$$\ln(|\Gamma|) = -2(L-z)/l_{gas} + \ln[R_2(1+R_1)/2]$$
$$\arg(\Gamma) = -2(L-z)/l_{gas} \qquad (7)$$

Note that both the logarithm of amplitude and the phase of the reflection coefficient have the same linear behaviour in the three variable *L, z* and $\sqrt{f}$. The introduction of a new degree of freedom allows to increase the number of methods useful for thermal diffusivity measurements. However the method we want to describe consists in calculating the reflection coefficient as a function of the cavity length *L* which can be varied by moving only the second mirror (*z* and $\sqrt{f}$ are constant). By the linear slopes of both phase and logarithm of amplitude of Γ, the diffusivity can be worked out by one of the two relationships

$$D_{gas} = 4\pi f\left(N_L/\frac{\Delta\ln(|\Gamma|)}{\Delta L}\right)^2 \qquad (8a)$$

$$D_{gas} = 4\pi f\left(N_L/\frac{\Delta\arg(\Gamma)}{\Delta L}\right)^2 \qquad (8b)$$

As usual the corrective factor N_L has been introduced to consider the effect of *a*. In figure 4 the result of a numerical study on the factor N_L is presented. The N_L values for both amplitude (see Eq.8a) and phase (see Eq.8b) are plotted for different diffusivity ratios D_{gas}/D_s (0.2 - 0.5 - 5) as a function of the parameter a/l_{gas}, which remains constant during the whole measurement. For large a/l_{gas} all the curves tend to 1 but in different ways so that the amplitude formula in Eq.(8a) has to be preferred already for a/l_{gas} >2. From a comparison between the 3 methods we have discussed we can conclude that there is a great analogy between the different formula for D_{gas} (see Eqs.4,5,8); in fact in all cases they are given by the product of three terms: the slope of the experimental data vs the variable chosen for the scan, the other variable kept constant, and a corrective factor *N*. Concerning the accuracy, the spatial scan methods (n°2, n°3), as discussed above, give rise to better results with respect to the frequency scan ones. Finally the use of a thermal wave resonator doesn't improve the corrective

factor (N_z and N_L tend to 1 in a similar way) but, due to the deflection gain, the accuracy on the experimental data and hence on the calculated slope is strongly increased. In case of a gas with nonhomogeneous diffusivity it is worth to note that each method can give, depending on its own philosophy on which is based, different aspects of the gas thermal diffusivity. In fact the first method guarantees an effective local diffusivity measurement, the second gives rise to the average diffusivity from the sample to the probe beam , while the third, by using the interference in the cavity, measures the average diffusivity in the whole resonator.

4. EXPERIMENTAL RESULTS

The cavity length scan method has been applied to measure the air thermal diffusivity inside the plane open resonator of fig.1. The scan has been performed moving the second mirror, a 20μm thick aluminium foil so to adjust the cavity length L from 100μm to more than 1300μm . The probe beam has been placed at about 70μm from the first mirror, the silicon coated glass sample which has been illuminated by a 250mW Ar laser with a spot on the silicon film of about 1mm. The mechanical chopper frequency was chosen f=36Hz so to have $a/l_{gas} \cong 2.3$.

The photothermal signal should be normalised to the reference signal which corresponds to an infinite cavity length. In practice one can use as a reference the photothermal signal for the maximum cavity length of 1300 μm. In fact such distance already inhibits the thermal waves from doing a complete round trip in the resonator. In figure 5 both phase and natural logarithm of amplitude are plotted vs the cavity length. Looking at the logarithm of amplitude one may note that within the first 300 μm (just one half of the air diffusion length) there is a gain for the deflection signal which reaches its maximum value of 11db. The reason of this low gain is due to radial losses. By applying Eqs.(7) the reflection coefficient Γ is carried out. In figure 6 both phase and natural logarithm of amplitude of Γ are plotted vs the cavity length L. The expected linear behaviour lasts till the first 500 μm, where the interference between the damped

forward and backward waves becomes ineffective. Looking at the linear slopes and using Eqs.(8) one should calculate the air thermal diffusivity. In this case a small distortion from linearity occurs to both phase and logarithm of amplitude. This effect can be related to a variable value of the air diffusivity. In fact with 250 mW pump beam power, by changing the cavity length of the resonator the d.c. temperature rise inside could change by several tens of degrees which are able to increase the air thermal diffusivity of several percent. By applying Eqs(8) locally, one obtains the calculated profile of air thermal diffusivity as a function of the cavity length (see fig.7). For cavity lengths larger than 450μm the effect of the d.c. temperature rise becomes negligible so that the diffusivity levels to the standard value of $0.21 cm^2/s$ at the room temperature of 20 C [5].

5. CONCLUSIONS

The theory of the plane thermal wave resonator is introduced for a gas cavity. The use of such device in order to perform accurate gas thermal diffusivity measurement is justified. Finally the experimental results on an air thermal resonator confirm the theory and give an example on how to calculate the air thermal diffusivity profile.

REFERENCES

1 C.A.Bennett Jr. and R.R.Patty, *Appl.Opt.* **21:** 49 (1982).

2 Jun Shen and Andreas Mandelis, *Rev.Sci.Instrum,* **66:** 4999 (1995).

3 M.Bertolotti, R.Li Voti, C.Sibilia, G.L.Liakhou, *SPIE* **2775:** 370 (1996).

4 M.Bertolotti, M. Firpo, R. Li Voti, S. Paoloni, C. Sibilia, F.Tani, G.L.Liakhou in *Progress in natural science,* Vol.6 219 (1996).

5 Y.S. Touloukian, R.W.Powell, C.Y. Yo and M.C.Nicolaou, in *Thermophysical Properties of Matter*, Vol.10 *Thermal diffusivity*, Plenum Press (1973).

CAPTION FOR FIGURES

Figure 1: Schematic representation of a thermal-wave resonator cavity; in situ investigation by photothermal deflection technique.

Figure 2: Numerical analysis of the factor N_f due to the finite spot size of the pump beam. In abscissa is *a/z*. The curves refer to different formula (phase and amplitude) and to different thermal diffusivity ratios between gas and sample *D*: amplitude formula; (curve 1) D=0.2; (curve 2) D=0.5; (curve 3) D=5; phase formula; (curve 4) D=0.2; (curve 5) D=0.5; (curve 6) D=5;

Figure 3: Numerical analysis of the factor N_z due to the finite spot size of the pump beam. In abscissa is a/l_{gas}. The curves refer to different formula (phase and amplitude) and to different thermal diffusivity ratios between gas and sample *D*: amplitude formula; (curve 1) D=0.2; (curve 2) D=0.5; (curve 3) D=5; phase formula; (curve 4) D=0.2; (curve 5) D=0.5; (curve 6) D=5;

Figure 4: Numerical analysis of the factor N_L , due to the finite spot size of the pump beam. In abscissa is a/l_{gas}. The curves refer to different formula (phase and amplitude) and to different thermal diffusivity ratios between gas and sample *D*: amplitude formula; (curve 1) D=0.2; (curve 2) D=0.5; (curve 3) D=5; phase formula; (curve 4) D=0.2; (curve 5) D=0.5; (curve 6) D=5;

Figure 5: Phase (radian) and natural logarithm of amplitude of the deflection signal as a function of the cavity length L (mm). The frequency is fixed to f=36Hz, The spot size is 1mm. The thermal wave resonator works in air.

Figure 6: Phase (radian) and natural logarithm of amplitude for the reflection coefficient Γ as a function of the cavity length L (mm). The data are obtained by using Eq.(7)

Figure 7: Air thermal diffusivity measurements vs the cavity length L (mm).

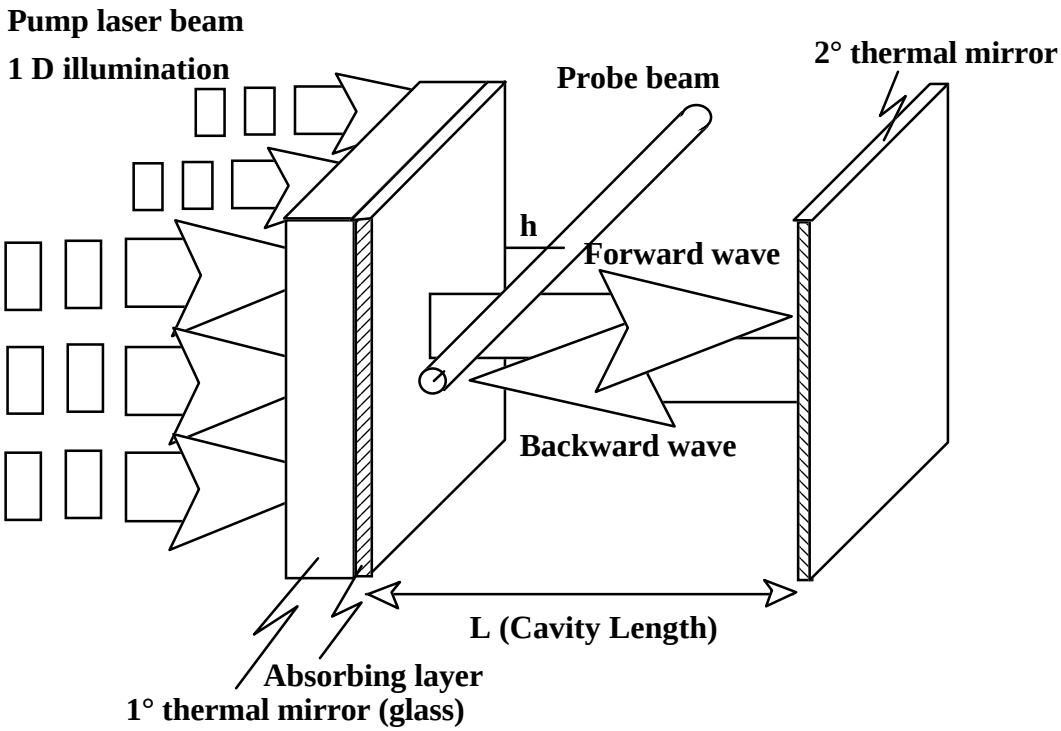
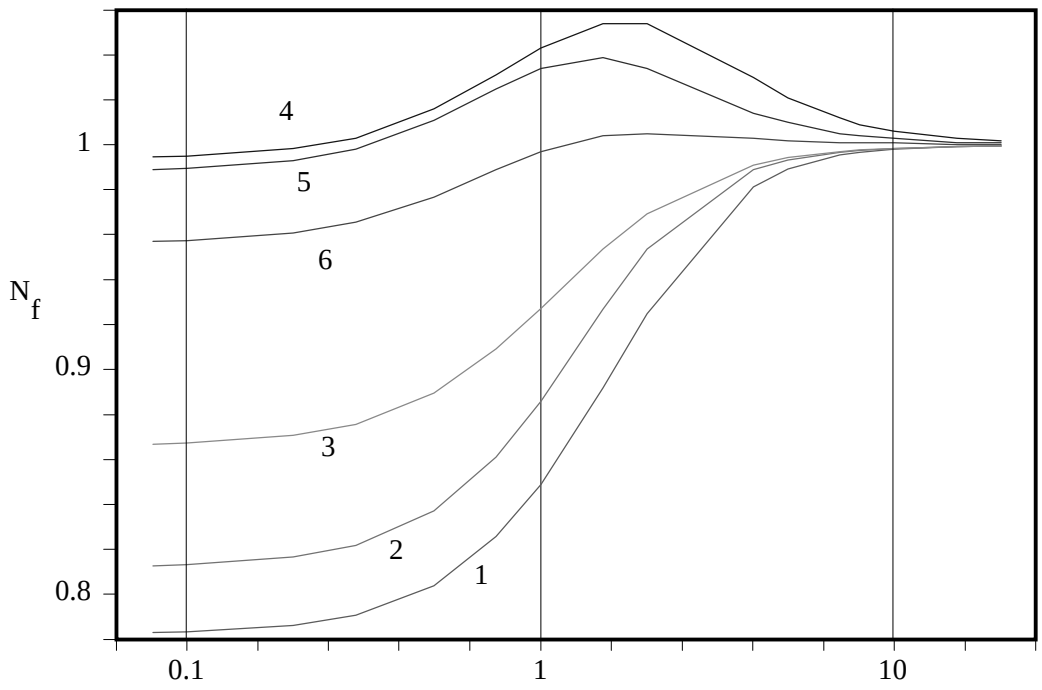


Figure 1



Spot normalised to the vertical offset

Figure 2

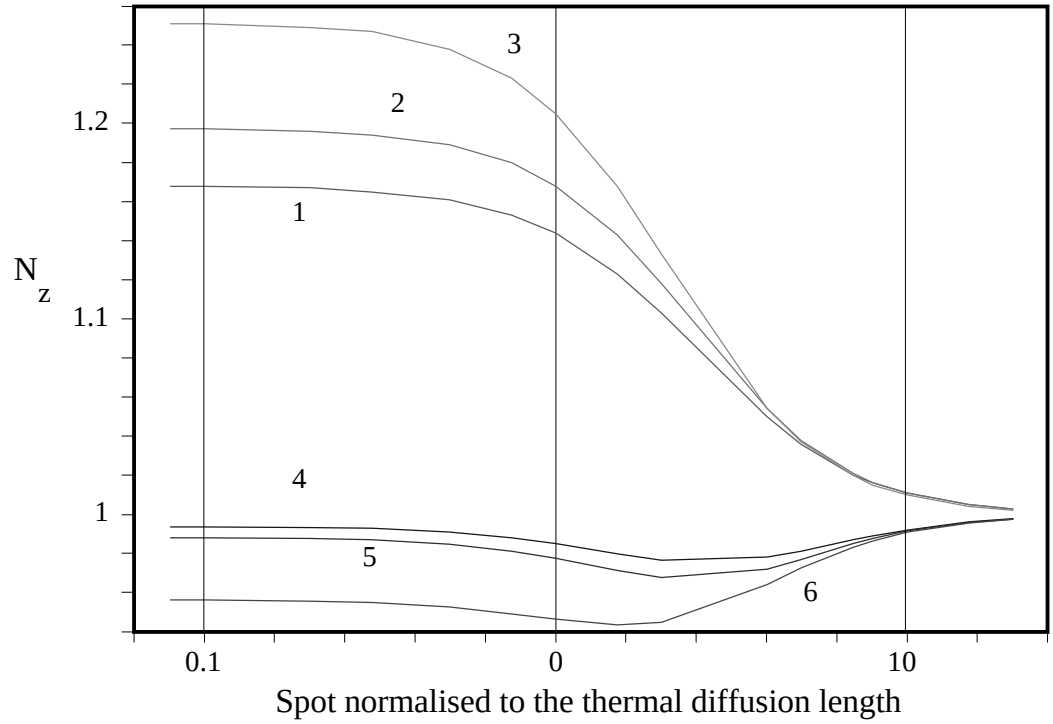


Figure 3

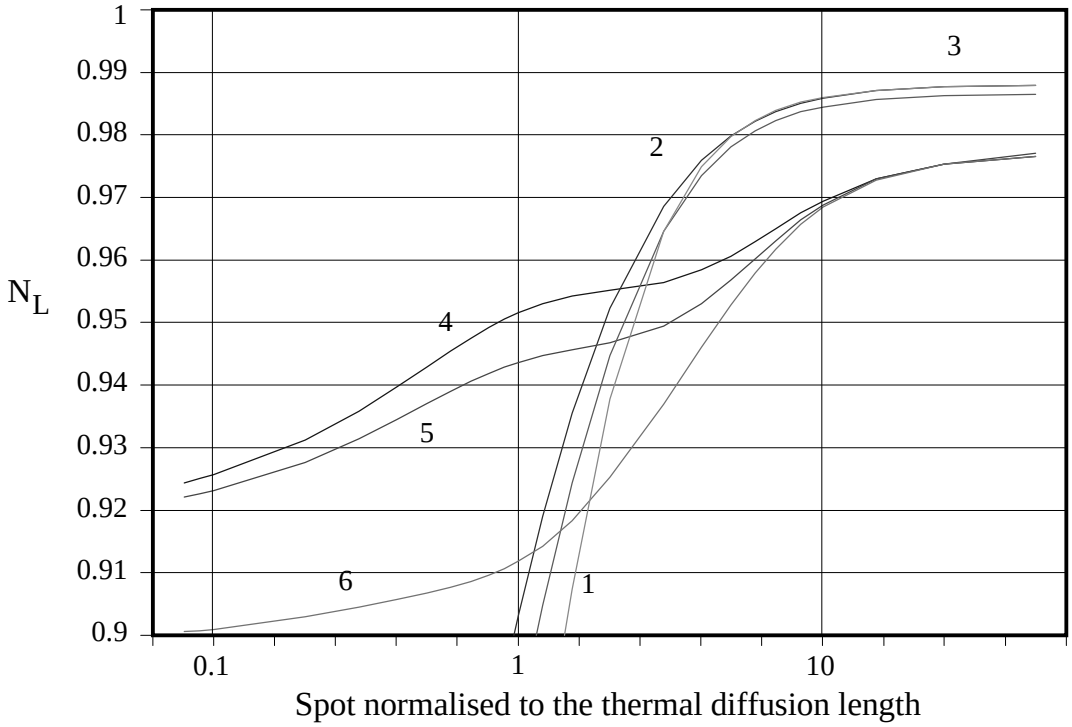


Figure 4

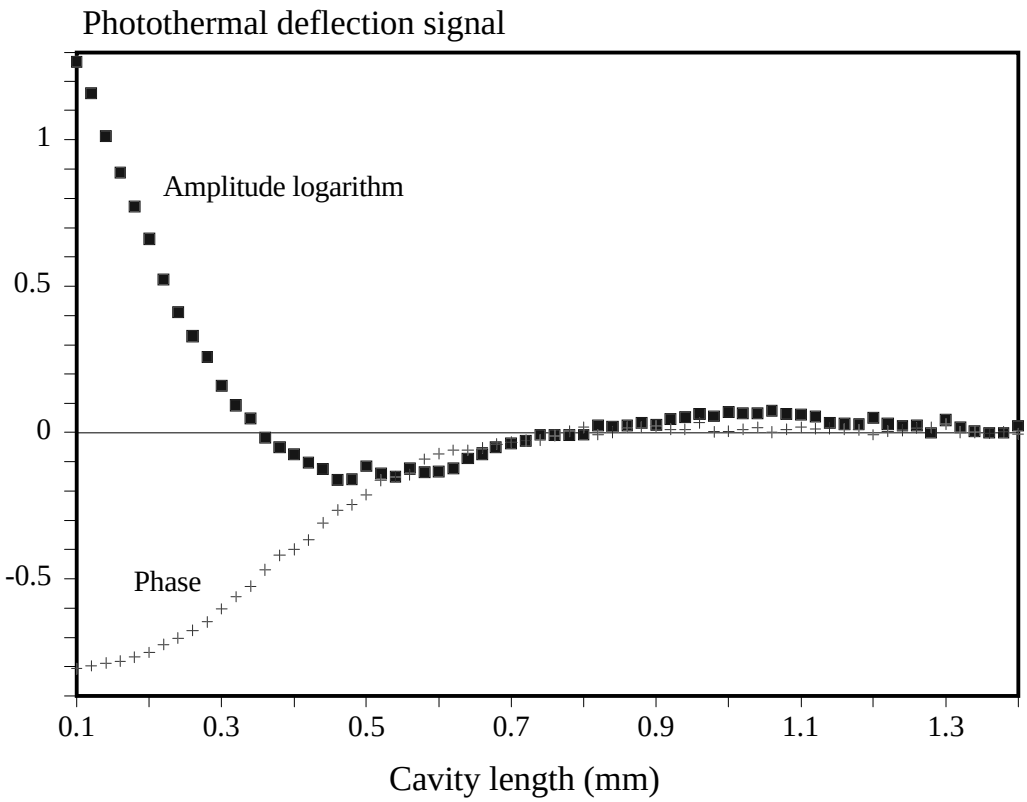


Figure 5

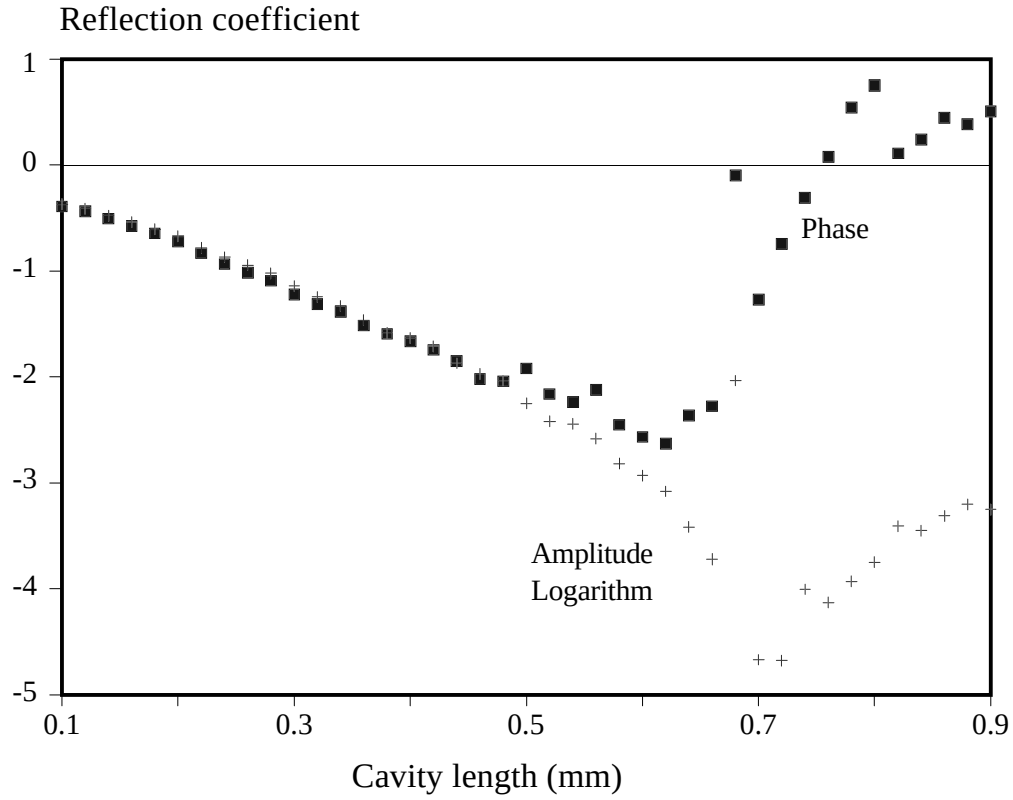


Figure 6

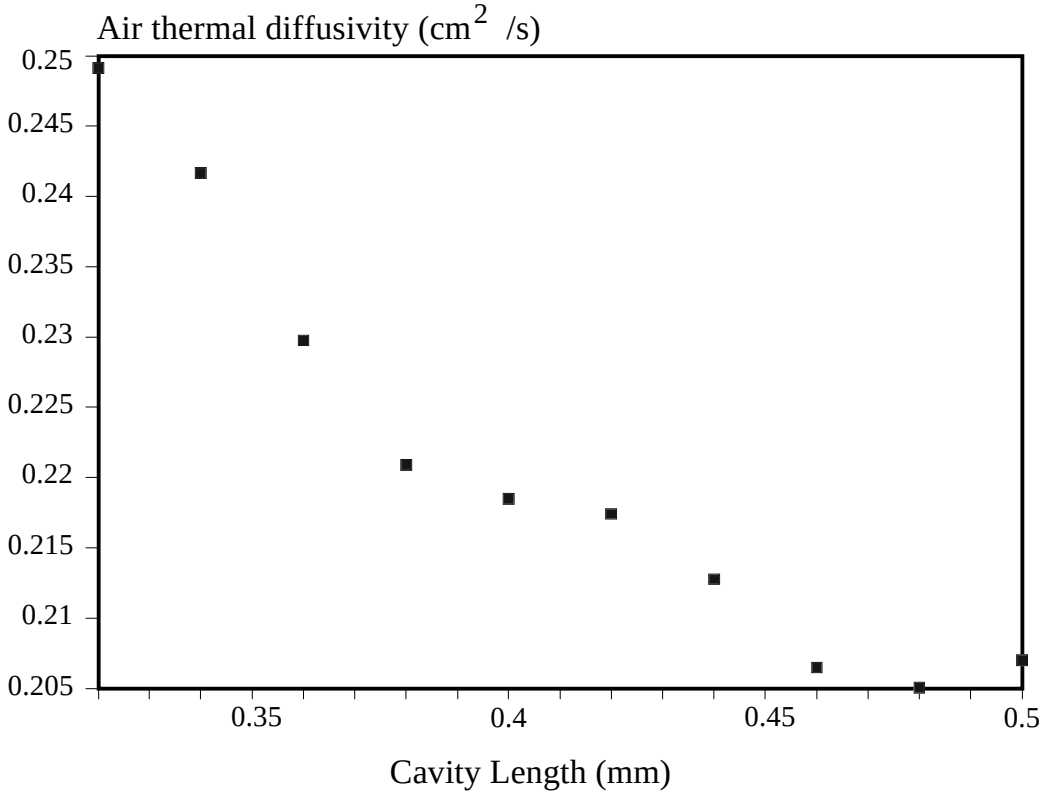

Air thermal diffusivity (cm2 /s)
0.25
0.245
0.24
0.235
0.23
0.225
0.22
0.215
0.21
0.205
0.35
0.4
0.45
0.5
Cavity Length (mm)

Figure 7